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Single-crystal growth and superconducting state of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$

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Abstract

Single crystals of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ were grown by a CsCl-flux method. Electrical resistivity $\rho(T)$ measurements were performed to reveal its superconducting properties. The $\rho(T)$ of the single crystal shows semiconducting-like behavior and superconducting transition at 3.4 K. The value is slightly higher than that of the polycrystalline sample with substitution amounts of $x = 0.2$ ($T_c \sim 2.5$ K). From $\rho(T)$ measurements in several magnetic fields, $\mu_0 H_{c2}^{//ab}(0)$ and $\mu_0 H_{c2}^{//c}(0)$ are estimated to be 17.2 T and 0.59 T, respectively. The superconducting anisotropic parameter γ is determined to be 29.2.

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Keywords: superconducto; BiS_2-based compound

1. Introduction

Since the discovery of superconductivity with superconducting transition temperatuure T_c at 8.6 K in the layered bismuth oxysulfide $Bi_4O_4S_3$ [1], other BiS_2-based superconductivity have been studied. $LaOBiS_2$ crystallizes with a space group $P4/nmm$ (No. 129) and this structure is composed of alternating superconducting BiS_2 layers and blocking LaO layers, which are similar to Fe-based superconductors. Substituting F for O induces superconductivity with $T_c = 2.5$ K [2], as well as tetravalent elements (Ti, Zr, Hf and Th) for La [3]. In addition, by replacing S atoms

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to Se atoms, T_c is enhanced: T_c = 4.2 K for $LaO_{0.5}F_{0.5}BiSSe$ [4], 3.7 K for $LaO_{0.5}F_{0.5}BiSe_2$ [5]. The enhancement of T_c in Se doped compounds is induced by an in-plane chemical pressure [6].

In this article, we report the single crystal growth of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ and its superconducting properties. In order to clarify superconducting state, we measured the electrical transport properties of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ at low temperature and strong magnetic fields up to 11 T.

2. Experimental Details

Single crystals of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ were grown by a CsCl-flux method similar to the previous reports [5,6]. The starting materials of 0.8 g and CsCl of 5 g were mixed and sealed in an evacuated quartz tube. The tube was heated up to 880 °C in 10 h and to 900 °C in 2 h, then kept for 10 h and cooled down to 650 °C for 125 h. The obtained materials were washed by water, ethanol, and acetone in order to remove the flux materials. The obtained single crystals were plate-like shape with approximately 1.0 × 1.0 mm^2. The crystal structure of the single crystal was examined by an X-ray diffraction (XRD) method using a conventional X-ray spectrometer equipped with Cu-Kα radiation and a monochromator (RAD-2X, Rigaku). Electrical resistivity $\rho(T)$ from 0.5 K to 300 K was measured under magnetic fields up to 11 T along the *ab*-plane and *c*-plane by dc-four-probe method in a ^3He cryostat (Oxford Heliox VL).

3. Experimental Result

3.1. X-ray diffraction

Figure 1 shows the XRD diffraction pattern of several single crystals $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ at room temperature. Only (00*l*) diffraction peaks were observed, indicating the crystallographic *c*-axis is perpendicular to the crystal plane. All the reflections can be indexed as the space group of *P*4/nmm and no extra peaks due to impurity phase can be detected from the XRD pattern. To estimate lattice constants, powder X-ray diffraction was performed by using powder crashed from single crystals. The lattice constants of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ are obtained to be *a* = 4.0856 Å and *c* = 13.453 Å. These values are slightly larger than those of the single crystal $LaO_{0.5}F_{0.5}BiS_2$, which suggest that S atoms are substituted with Se atoms.

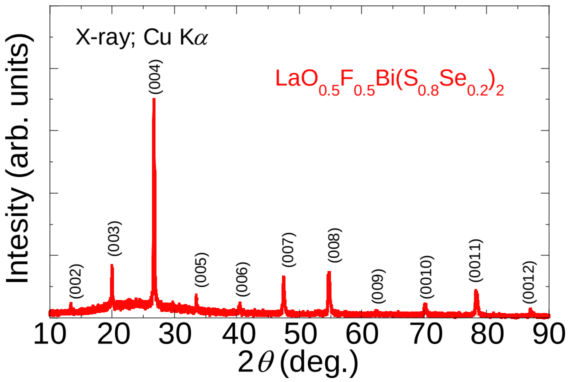

Fig. 1. XRD diffraction pattern of single crystals $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ at room temperature.

3-2. Electrical resistivity

Figure 2 shows temperature dependence of the electrical resistivity $\rho(T)$ of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ down to 0.5 K with electrical current along the *ab*-plane. A semiconducting-like behavior was observed and superconducting transition was detected. T_c is defined as 50 % drop from residual resistivity ρ_0 and determined to be 3.4 K. ρ_0 is defined at just above T_c (ρ_0 = 3.5 mΩcm). Compared with polycrystalline sample of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ [7], although the behavior of $\rho(T)$ and the ρ_0 is almost the same value, T_c is slightly higher than that of polycrystalline sample (T_c ~ 2.5 K).

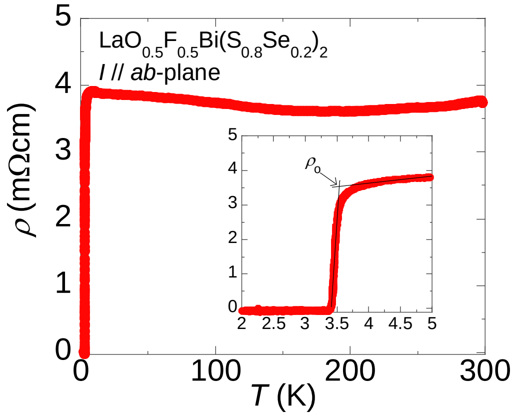

Fig. 2. Temperature dependence of the electrical resistivity $\rho(T)$ of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$. The Inset shows $\rho(T)$ near T_c.

Figure 3 shows temperature dependence of electrical resistivity $\rho(T)$ of single crystal $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ in several magnetic fields along *ab*-plane (a) and *c*-plane (b). T_c shifts toward lower temperature with an increase of the magnetic field. Superconductivity is robust against magnetic field applying along *ab*-plane whereas it is quite sensitive to the magnetic field applying along *c*-plane.

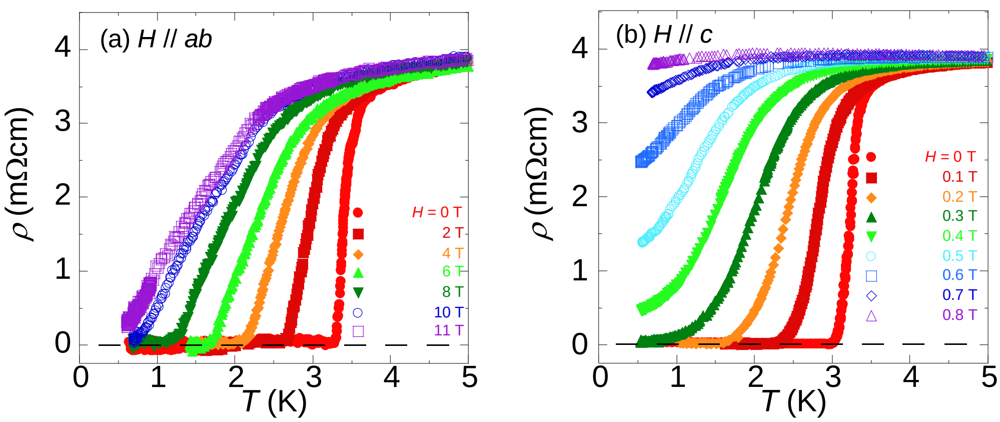

Fig. 3. Temperature dependence of the electrical resistivity $\rho(T)$ of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ in several magnetic fields along *ab*-plane (a) and *c*-plane (b) at low temperatures.

Figures 4(a) and (b) show temperature dependence of upper critical field $\mu_0 H_{c2}^{//ab}(T)$ and $\mu_0 H_{c2}^{//c}(T)$. Error bars are determined by the temperature of 10 % and 90 % drop from ρ_0. For *ab*-plane, the $\mu_0 H_{c2}^{//ab}(T)$ curve shows non-*T*-liner behavior. A Werthamer-Helfand-Hohenberg (WHH) fitting in a dirty limit, as presented by dashed line, is not completely correspond to experimental data. The type-II SC approximation $\mu_0 H_{c2}(T) = \mu_0 H_{c2}(0)(1-(T/T_c)^{3/2})^{3/2}$ [8], presented by solid line, is in good agreement with the data. $\mu_0 H_{c2}^{//ab}(0)$ is estimated to be 17.2 T. For *c*-plane, the $\mu_0 H_{c2}^{//c}(T)$ curve shows nearly *T*-linear behavior. Because the WHH fitting and type-II SC approximation are not corresponding to the data, we fitted $\mu_0 H_{c2}^{//c}(T)$ using polynomial equation $\mu_0 H_{c2}(T) = \sum_{n=0}^{3} a_n T^n$ presented by solid line. $\mu_0 H_{c2}^{//c}(0)$ is estimated to be 0.59 T. The superconducting anisotropic parameter $\gamma = H_{c2}^{//ab}(0)/H_{c2}^{//c}(0)$ is determined to be 29.

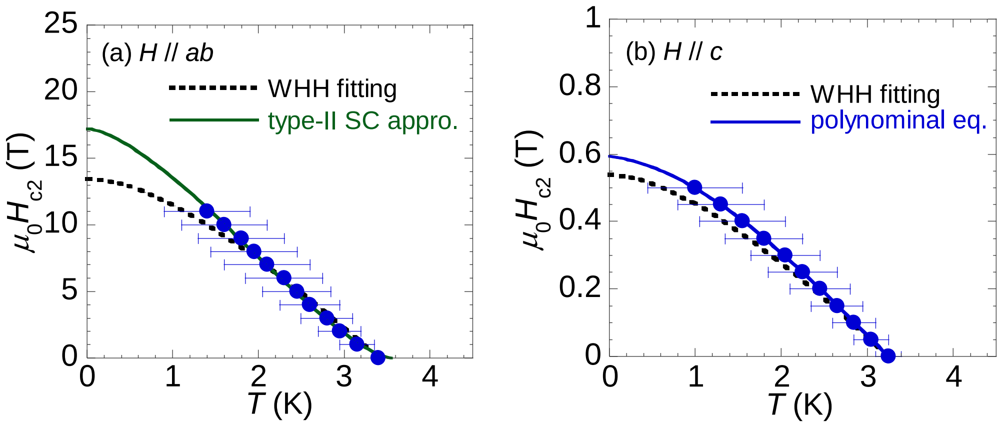

Fig. 4(a) Temperature dependence of the upper critical field $\mu_0 H_{c2}(T)$ of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ in several magnetic fields along *ab*-plane. The dotted line shows the WHH prediction in a dirty limit. The Solid line represents type-II SC approximation. (b). Temperature dependence of $\mu_0 H_{c2}(T)$ along *c*-plane. The dotted line shows the WHH prediction in a dirty limit. Solid line represents polynomial equation.

4. Conclusion

We succeeded in growing the single crystal of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$ by a CsCl-flux method. The single crystal sample exhibits superconducting transition at 3.4 K and has strong anisotropy. $\mu_0 H_{c2}^{//ab}(T)$ and $\mu_0 H_{c2}^{//c}(T)$ curves cannot be fitted well by the WHH theory. We employed the type-II SC approximation for *ab*-plane and polynomial equation for *c*-plane. The estimated values of $\mu_0 H_{c2}^{//ab}(0)$ and $\mu_0 H_{c2}^{//c}(0)$ are 17.2 T and 0.59 T, respectively. The superconducting anisotropic parameter γ is determined to be 29.2.

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